# PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

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### INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/EP2004/012514

	Вох	No. I Basis of the report						
1.		With regard to the language, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.						
	$\square$ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:							
		<ul> <li>□ international search (under Rules 12.3 and 23.1(b))</li> <li>□ publication of the international application (under Rule 12.4)</li> <li>□ international preliminary examination (under Rules 55.2 and/or 55.3)</li> </ul>						
2.	With regard to the <b>elements*</b> of the international application, this report is based on <i>(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report):</i>							
	Des	cription, Pages						
	1-19	as originally filed						
	Clai	ms, Numbers						
	1-18	received on 07.09.2005 with letter of 06.09.2005						
		a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing						
3.		☐ The amendments have resulted in the cancellation of:						
		<ul><li>☐ the description, pages</li><li>☐ the claims, Nos.</li></ul>						
	-	☐ the drawings, sheets/figs						
		☐ the sequence listing (specify): ☐ any table(s) related to sequence listing (specify):						
4.		This report has been established as if (some of) the amendments annexed to this report and listed below not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the plemental Box (Rule 70.2(c)).						
		<ul> <li>□ the description, pages</li> <li>□ the claims, Nos.</li> <li>□ the drawings, sheets/figs</li> </ul>						
		<ul> <li>□ the sequence listing (specify):</li> <li>□ any table(s) related to sequence listing (specify):</li> </ul>						
	*	If item 4 applies, some or all of these sheets may be marked "superseded."						

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/EP2004/012514

		No. Ill Non-establishment of the licability	of opi	nion with regard to novelty, inventive step and industrial	
1.		The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:			
		the entire international applicat	ion,		
☑ claims Nos. 1-4, and 9-17 (all partially), 18 (completely)			lly), 18 (completely)		
because:					
		the said international application not require an international pre		the said claims Nos. relate to the following subject matter which does ary examination (specify):	
	the description, claims or drawings (indicate particular elements below) or said claims Nos. are so uncleate that no meaningful opinion could be formed (specify):				
		the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opin could be formed.			
no international search report has been established for the said claims Nos. 1-4, and 9-17 (all partial (completely)  the nucleotide and/or amino acid sequence listing does not comply with the standard provided for in C of the Administrative Instructions in that:			een established for the said claims Nos. 1-4, and 9-17 (all partially), 18		
		the written form		has not been furnished	
				does not comply with the standard	
		the computer readable form		has not been furnished	
				does not comply with the standard	
				and/or amino acid sequence listing, if in computer readable form only, do ements provided for in Annex C-bis of the Administrative Instructions.	
	$\boxtimes$	See separate sheet for further	detai	ils	

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International application No. PCT/EP2004/012514

_	Box	k No. IV La	ck of unity of inv	ention			
1.							
2.		☐ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.					
3.	Thi	This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is					
		complied with.					
	$\boxtimes$	not complied with for the following reasons:					
		see separate sheet					
4.	Co	Consequently, this report has been established in respect of the following parts of the international application:					
		all parts.					
	$\boxtimes$	the parts relating to claims Nos. 1-4, and 9-17 (all partially, namely as far as they concern invention 1).					
_			easoned stateme itations and expla				ard to novelty, inventive step or industrial tement
1.	Sta	atement					
	Novelty (N)		Yes: Claims No: Claims		4, 14-17 1-3, 9-13	•	
	Inventive step (IS)		Yes: No:	Claims Claims	4 1-3, 9-17		
	Inc	lustrial applic	ability (IA)	Yes: No:	Claims Claims	1-4, 9-17	
2	Cit	eations and ex	volanations (Rule 3	70.71.			

see separate sheet

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

#### Re Item III.

The International Searching Authority had raised an objection of lack of unity of invention and defined 10 different alleged inventions (see below re item IV). Upon invitation to pay additional search fees according to Article 17(3)(a) and Rule 40.1 PCT (Form PCT/ISA/206), no required additional search fees were timely paid by the applicant. Consequently the International Search Report was restricted to the invention first mentioned in the claims, which invention is covered by part of the subject-matter of claims 1-4, and 9-17.

Consequently also this report is restricted to the subject-matter for which an International Search Report was established, i.e. said first alleged invention as it is defined in the amended claims.

#### Re Item IV.

In response to the first written opinion, i.e. the Written Opinion of the International Searching Authority (WO-ISA) being considered to be a written opinion of the International Preliminary Examining Authority (IPEA), the applicant filed amendments. In the first claim [under b), first paragraph] it reads now instead of "... an ethylene modulator, which inhibits the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC), such as derivatives of vinylglycine, hydroxylamines, oxime ether derivatives", which latter phrase had no limiting effect, "... (ACC), selected from derivatives of vinylglycine and hydroxylamines". This amendment, evidently intended to limit the scope of the first alleged invention, leaves the remaining subject-matter, i.e. the other alleged inventions, unchanged.

Consequently the considerations concerning unity of invention remain substantially the same as for the set of claims originally filed, which were already communicated with the first written opinion. Only the definition of the first item needs to be modified in conformity with the amendment.

The separate inventions are:

1. covered by parts of the subject-matter of claims 1-4, and 9-17
A mixture of strobilurin type fungicides of formula (I) and an ethylene modulator, which inhibits the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic

acid (ACC), selected from derivatives of vinylglycine and hydroxylamines, and corresponding processes

- 2. covered by parts of the subject-matter of claims 1-4, and 9-17, and the subject-matter of claims 5 and 8
- A mixture of strobilurin type fungicides of formula (I) and Co<sup>++</sup> or Ni<sup>++</sup> ions in plant available form as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 3. covered by parts of the subject-matter of claims 1-3, and 9-17 A mixture of strobilurin type fungicides of formula (I) and phenolic scavengers such as n-propyl gallate as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 4. covered by parts of the subject-matter of claims 1-3, and 9-17
  A mixture of strobilurin type fungicides of formula (I) and polyamines such as putrescine, spermine or spermidine as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 5. covered by parts of the subject-matter of claims 1-4, and 9-17
  A mixture of strobilurin type fungicides of formula (I) and structural analogs of ACC, such as -aminoisobutyric acid or L-aminocyclopropene-1-carboxylic acid as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 6. covered by parts of the subject-matter of claims 1-4, and 9-17, and the subject-matter of claim 7
- 6.1 A mixture of strobilurin type fungicides of formula (I) and salicylic acid or acibenzolar-S-methyl as ethylene modulator, which block the conversion of ACC into ethylene, and a process for controlling rust in legumes by applying said mixture,
  - 6.2 a process for increasing yield and quality of legumes by applying said mixture, 6.3 a process for reducing ethylene evolution of plants by applying said mixture, and 6.4 a process for reducing undesired defoliation of crop plants by applying said mixture.

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

7. covered by parts of the subject-matter of claims 1-4, and 9-17, and the subject-matter of claims 6 and 8

A mixture of strobilurin type fungicides of formula (I) and structure analogs of ascorbic acid which act as inhibitors of ACC oxidase, such as prohexadione-Ca or trinexapac-ethyl as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes

- 8. covered by parts of the subject-matter of claims 1-3, and 9-17
- 8.1 A mixture of strobilurin type fungicides of formula (I) and triazolyl compounds such as paclobutrazole or uniconazole, as inhibitors of cytochrome P-450-dependent monoxygenases whose main action is to block the biosynthesis of gibberellins, as ethylene modulator, which block the conversion of ACC into ethylene, and a process for controlling rust in legumes by applying said mixture
  - 8.2 a process for increasing yield and quality of legumes by applying said mixture,
  - 8.3 a process for reducing ethylene evolution of plants by applying said mixture, and
  - 8.4 a process for reducing undesired defoliation of crop plants by applying said mixture.
- 9. covered by parts of the subject-matter of claims 1-4, and 9-17 A mixture of strobilurin type fungicides of formula (I) and an ethylene modulator, which inhibits the action of ethylene, and corresponding processes.
- 10. covered by the subject-matter of claim 18
  A method for controlling harmful plant pathogens by applying an effective amount of Co<sup>++</sup> ions in plant-available form.

They are not so linked as to form a single general inventive concept (Rule 13.1 PCT) for the following reasons:

Documents cited in this motivation are referred to as D1, D2 etc. in the order of their occurrence in the search report and as listed below under re item V. The disclosures of these documents referred to can be found in the passages cited in the search report.

I. The subject-matter of the claims as a whole defines solutions to the very general problem of providing agriculturally beneficial processes. Already from this starting point

there are two alternative solutions each characterised by the active agents used in the processes.

A first solution, directed to the more specific processes of controlling rust in legumes, increasing yield and quality of legumes, reducing ethylene evolution of plants and reducing undesired defoliation of crop plants respectively is characterised by applying a mixture of a strobilurin type fungicide of formula (I) and an ethylene modulator. A second solution, directed to a method of controlling harmful plant pathogens is characterised by applying Co<sup>++</sup> ions.

These different solution only share the common feature that they are solutions to the above defined very general technical problem. Evidently this general problem and solutions to it are known, for instance all the cited documents are concerned with providing such solutions. This common feature can therefore not be a special technical feature in the sense of Rule 13.2 PCT. Consequently the claimed subject-matter is not so linked as to form a single general inventive concept, i.e. it lacks unity of invention in the sense of Rule 13.1 PCT.

In a first step the claimed subject-matter has therefore to be subdivided into two alleged invention.

- 1. Covered by the subject-matter of claims 1-17: Agriculturally beneficial processes, as specified in claims 11-17, characterised by the application of a mixture of a strobilurin type fungicide of formula (I) and an ethylene modulator, and the corresponding mixtures.
- 2. Covered by the subject-matter of claims 18: A method for controlling harmful plant pathogens by applying an effective amount of Co<sup>++</sup> ions in plant-available form.
- II. Concerning item 1 three alternative solutions are proposed: The ethylene modulator
- a) belongs to certain classes of compounds which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC),
- b) blocks the conversion of ACC into ethylene
- c) inhibits the action of ethylene.

D1 discloses mixtures of pyraclostrobin with kresoxim-methyl or epoxiconazol. D2 refers to commercially available binary and ternary mixtures comprising such mixtures, and D3 also

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

discloses mixtures of pyraclostrobin and epoxiconazol. From D3, D7 and D8 it is known that pyraclostrobin and kresoxim-methyl are ethylene modulators inhibiting the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid, while epoxiconazole is known from D3 to block the conversion of ACC into ethylene. D4 discloses mixtures of the strobilurin-type fungicide orysastrobin, with the ethylene modulator acibenzolar-S-methyl, both compounds are also specified in the present application. Further D5 and D6 also propose combinations of strobilurin type fungicides with the ethylene modulators paclobutrazole and uniconazole, all compounds are also specified in the present application. These ethylene modulators act by blocking the conversion of ACC into ethylene.

Therefore the common feature of the three solutions of item 1, namely that the second component of the strobilurin type fungicides containing mixtures is an ethylene modulator, since it is already known from the prior art, cannot function as a special technical feature in the sense of Rule 13.2 PCT either.

Consequently in a second step the subject-matter of claims 1-17 has to be further subdivided into three alleged invention:

- 1.1 A mixture of strobilurin type fungicides of formula (I) and an ethylene modulator, which inhibits the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC), selected from derivatives of vinylglycine and hydroxylamines, and corresponding processes.
- 1.2 A mixture of strobilurin type fungicides of formula (I) and an ethylene modulator, which blocks the conversion of ACC into ethylene, and corresponding processes
- 1.3. A mixture of strobilurin type fungicides of formula (I) and an ethylene modulator, which inhibits the action of ethylene, and corresponding processes.
- III. D1 by disclosing combinations of kresoxim-methyl and pyraclostrobin both being at the same time strobilurin type fungicides of formula (I) and ethylene modulators, which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid is novelty destroying for the subject-matter of item 1.1 (see below under re item V). The classes of structurally defined ethylene modulators specified in the claim for item 1.1 are not defined precisely enough to delimitate them from some of the strobilurin type fungicides, like e.g. kresoxim-methyl (see below under re item V).

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

However, it is evident on closer investigation that the more specific classes of modulators mentioned in claim 4 and on page 5, lines 24-31 (e.g. aminoethoxy-vinylglycine and aminooxyacetic acid) share not only said mode of action and the general structural feature of being derivatives of vinylglycine or hydroxylamines, but also some common, although not explicitly defined, structural feature, which is lacking in strobilurin type ethylene modulators like kresoxim-methyl. Therefore there is unity of invention within this more specific and novel subject-matter characterised by these structurally more precisely defined ethylene modulators.

The situation is different for the subject-matter of item 1.2, which is characterised by the common general feature that the ethylene modulator blocks the conversion of ACC into ethylene, and a series of alternative additional features further characterising the ethylene modulators structurally.

Since the cited prior art of D1-D6 already discloses (see above) combinations of strobilurin type fungicides with ethylene modulators, which block the conversion of ACC into ethylene, the general feature common to the subject-matter of item 1.2 cannot be a single special technical feature indicating unity of the subject-matter of item 1.2.

The classes of modulators seem not to share beside the mode of action any more specific structurally defined feature. In fact, even if there was a further common, although not explicitly defined, structural feature, it would necessarily already be present in the cited prior art of D4-D6, since these documents disclose combinations with modulators explicitly mentioned among the modulators of item 1.2, namely acibenzolar-S-methyl, paclobutrazole and uniconazole.

Therefore the subject-matter of item 1.2 has to be further subdivided into 7 different alleged inventions:

- 1.2.1 A mixture of strobilurin type fungicides of formula (I) and Co<sup>++</sup> or Ni<sup>++</sup> ions in plant available form as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 1.2.2 A mixture of strobilurin type fungicides of formula (I) and phenolic scavengers such as n-propyl gallate as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

- 1.2.3 A mixture of strobilurin type fungicides of formula (I) and polyamines such as putrescine, spermine or spermidine as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 1.2.4 A mixture of strobilurin type fungicides of formula (I) and structural analogs of ACC, such as -aminoisobutyric acid or L-aminocyclopropene-1-carboxylic acid as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 1.2.5 A mixture of strobilurin type fungicides of formula (I) and salicylic acid or acibenzolar-S-methyl as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 1.2.6 A mixture of strobilurin type fungicides of formula (I) and structure analogs of ascorbic acid which act as inhibitors of ACC oxidase, such as prohexadione-Ca or trinexapac-ethyl as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- 1.2.7 A mixture of strobilurin type fungicides of formula (I) and triazolyl compounds such as paclobutrazole or uniconazole, as inhibitors of cytochrome P-450-dependent monooxygenases whose main action is to block the biosynthesis of gibberellins, as ethylene modulator, which block the conversion of ACC into ethylene, and corresponding processes
- IV. Following the above scheme one arrives after renumeration at the list of 10 alleged inventions as defined above, thereby subdividing the claimed subject-matter into what the IPEA Authority believes to be the minimum number of alleged inventions.

#### Re Item V.

The following opinion is for the reasons given under re item III restricted to those parts of the subject-matter of the claims relating to the first invention as it was defined by the International Searching Authority and again by this International Examining Authority under re item IV.

The following documents are referred to in this communication:

PCT/EP2004/012514

D1: WO 97/40688 A

D2: TOMLIN C (ED): "The e-Pesticide Manual 2003 (Thirteenth Edition) Version 3.0" November 2003 (2003-11), BRITISH CROP PROTECTION COUNCIL, LONDON, GB; entry "PYRACLOSTROBIN", ISSN: 0306-3941

D3: JABS, T. ET AL: "Anti-oxidative and anti-senescence effects of the strobilurin pyraclostrobin in plants: A new strategy to cope with environmental stress in cereals" BCPC CONFERENCE--PESTS & DISEASES, 2002 (VOL. 2), 941-946

D4: WO 99/48370 A

D5: US 2003/060371 A1

D6: US-B1-6 369 090

D7: KOEHLE, H. ET AL: "Physiological effects of the new fungicide Jewel on yield in cereals" GESUNDE PFLANZEN, 49(8), 267-271, 1997,

D8: GROSSMANN, KLAUS ET AL: "Bioregulatory effects of the fungicidal strobilurin kresoxim-methyl in wheat (Triticum aestivum)" PESTICIDE SCIENCE, 50(1), 11-20, 1997

D9: WO 96/00005 A D10: US-A-5 869 424

# Clarity and Interpretation of the claims

The further definition of the ethylene modulators, which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC), as being "selected from derivatives of vinylglycine and hydroxylamines" introduced by amendment is not sufficiently clear (Article 6 PCT).

Based on claim 4 and the description page 5, lines 24-31, it is assumed that this should be read as 'selected from derivatives of vinylglycine and <u>derivatives of</u> hydroxylamine', after all e.g. aminooxyacetic acid is not (a) hydroxylamine.

Based on the fact that the third class specified in this context in originally filed claim 1, namely "oxime ether derivatives" is now omitted, it is assumed that what was intended by the amendment was the exclusion of the latter from the scope of the claim.

However, the term derivative itself is too vague to allow this delimitation. The aminooxyacetic acid mentioned in claim 4 is for instance apparently considered to be a derivative of hydroxylamine. This seems reasonable, because it can be seen as the result of the etherification of the hydroxyl group of hydroxylamine. Analogously an oxime can also be considered to be a hydroxylamine derivative because it can be seen as the result

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

of the derivatisation of the amino group of hydroxylamine. Consequently also an oxime ether as the result of both types of derivatisation has to be seen as falling under the definition 'derivative of hydroxylamine'.

Therefore claim 1 is interpreted as covering mixtures comprising as one component an oxime ether, which inhibits the conversion of S-adenosyl-L-methionine into 1-amino-cyclopropane-1-carboxylic acid (ACC), like for instance the strobilurin kresoxim-methyl.

## **Novelty**

The subject-matter of claims 1-3, 9-13 is not new (Article 33(1) und (2) PCT).

Independent claim 1 claims mixture comprising a strobilurin type fungicide of formula (I) of claim 1 and one or more ethylene modulator, which is an ethylene biosynthesis inhibitor which inhibits the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC), selected from derivatives of vinylglycine and hydroxylamines. The scope being interpreted as explained in the foregoing.

Independent claims 11, 14-17 (of which claims 14 and 15 are seen as virtually identical) are directed to processes for controlling rust in legumes, for increasing yield and quality of legumes, for reducing ethylene evolution of plants by applying said mixture, and for reducing undesired defoliation of crop plants by applying said mixture.

Depending claims add further feature concerning the strobilurin type fungicide being e.g. kresoxim-methyl, orysastrobin or pyraclostrobin (claims 2, and 3), the ethylene modulator, being e.g. aminoethoxyvinylglycine or aminooxyacetic acid (claim 4), additional presence of an azole type fungicide, being e.g. epoxiconazole (claim 9), or of a surfactant, being e.g. sodium dodecylsulphate (claim 10), that the rust infected legumes are soya plants (claim 12), and the pathogen is e.g. *Phakopsora pachyrhizi* (claim 13).

D1 discloses (see the passages cited in the search report) fungicidal compositions comprising the carbamate strobilurin pyraclostrobin, which is the most preferred strobilurin of the present application (claim 3) with an oximether strobilurin, e.g. kresoxim-methyl, which is one of the preferred strobilurins of the present application (claim 2), in particular for treating cereals like wheat, for instance against rust. A fungicidal triazole, like e.g. epoxiconazole, and surfactants like e.g. a fatty alcohol sulphate may be added.

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

These compositions are novelty destroying for claims 1-3, 9 and 10 since these strobilurins are ethylene modulating hydroxylamine derivatives, namely oximether, which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC) (see documents D3, D7 and D8).

From D2 it can be derived that ternary mixtures of pyraclostrobin, kresoxim-methyl and epoxiconazole are actually already commercially available under the tradename Opponent® and that pyraclostrobin is used for controlling *Phakopsora pachyrhizi* in soybeans. Therefore D2 is novelty destroying for claims 1-3, and 9-13.

The remaining cited documents are not novelty destroying for the claimed subject-matter. Therefore claims 4 and 14-17 are new (Article 33(1) and (2) PCT)

D3 discloses (see the passages cited in the search report) the anti-oxidative and anti-senescence effect of pyraclostrobin on cereals and its positive effects on yield even in the absence of fungal pathogens. It is shown to prevent the release of stress induced ethylene.

D4 discloses (see the passages cited in the search report) synergistic combinations of tripeloximether strobilurins, like e.g. orysastrobin with the resistance inducers probenazole or acibenzolar-S-methyl.

D5 discloses (see the passages cited in the search report) the increase of yield and vigour of plants, e.g. soybeans, by application of a combination of a strobilurin, e.g. azoxystrobin, and an azole, e.g. paclobutrazole.

D6 discloses (see the passages cited in the search report) ternary mixtures of pyrazol-strobilurins, e.g. pyraclostrobin or dimoxystrobin with fenpropimorph, tridemorph or fenpropidin and an azole, e.g. uniconazole.

D7 discloses (see the passages cited in the search report) synergistic physiological effects of kresoxim-methyl and epoxiconazole on cereals. which lead to an increase of yield even in the absence of fungal pathogens. It is taught that kresoxim-methyl prevents the release of stress induced ethylene due to ACC synthase inhibition while epoxiconazole does so due to the blocking of conversion of ACC into ethylene.

D8 discloses (see the passages cited in the search report) the effect of kresoxim-methyl on ethylene production, leaf senescence, and yield of cereals due to the inhibition of ACC synthase. The effects are compared with those of the ethylene biosynthesis inhibitor

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

aviglycine (=aminoethoxyvinylglycine), which also inhibits the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid,

D9 discloses (see the passages cited in the search report) a process for soil improvement by applying ethylene modulators, like e.g. the vinylglycines rhizobitoxin and aminoethoxyglycine, the hydroxylamine derivatives L-canaline and aminooxyacetic acids and certain known oxime ethers structurally related to these. This soil treatment promotes shoot and root biomass and root nodule formation in soybeans.

Finally D10 discloses (see the passages cited in the search report) combinations of plant growth regulators comprising an ethylene modulator, which inhibits ACC synthesis, preferably the oximethers mentioned above and a second growth regulator acting as a growth retardant, e.g. mepiquat.

## **Inventive step**

1. The subject-matter of claims 1-3, and 9-17 does not involve an inventive step (Article 33(1) and (3) PCT).

The subject-matter of claims 1-3, and 9-13 cannot involve an inventive step since it lacks novelty.

Using the combinations of D1 in processes according to claims 11-17, i.e. a process for controlling rust in legumes, for increasing yield and quality of legumes, for reducing ethylene evolution of plants, and for reducing undesired defoliation of crop plants was obvious in the light of the teachings of documents D2 and D3, and also D7 and D8. These documents already disclosed that pyraclostrobin and kresoxim-methyl respectively show such physiological effects.

2. The subject-matter of claim 4 involves an inventive step (Article 33(1) and (3) PCT).

In the light of the description and the closest prior art of D1 the problem underlying the invention can be seen in the provision of further compositions comprising a strobilurin type fungicide and a second agriculturally active agent, and corresponding agricultural processes using such compositions.

The solution of claim 4 is characterised by the combination of a strobilurin type fungicide of formula (I) of claim 1 and the ethylene modulator aminoethoxyvinylglycine or aminooxyacetic acid, which both are a ethylene biosynthesis inhibitors which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC).

# INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY (SEPARATE SHEET)

PCT/EP2004/012514

The closest prior art of D1 discloses compositions comprising two strobilurins, both being fungicides and at the same time ethylene biosynthesis inhibitors which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid. Strobilurins like pyraclostrobin are structurally unrelated to aminoethoxyvinylglycine and aminooxyacetic acid. Oximether strobilurins like kresoxim-methyl are only remotely related to the latter, by virtue of being hydroxylamine derivatives. Therefore the skilled person would not have expected, merely based on the fact that strobilurins have beside their fungicidal action a secondary effect on the ethylene production of plants, that one could replace one of the oximether-strobilurins of these combinations with these structurally very different ethylene modulators.

Also starting from D4 which discloses compositions comprising beside a strobilurin type fungicide the ethylene modulator acibenzolar-S-methyl, which acts by blocking the conversion of ACC into ethylene, or D5 or D6 which also combine with ethylene modulators of this type the subject-matter does not become obvious. It is not suggested in the prior art to replace the ethylene modulators used in D4-D6 by aminoethoxyvinylglycine or aminooxyacetic acid, which inhibit the conversion of S-adenosyl-L-methionine into 1-aminocyclo-propane-1-carboxylic acid, nor is it obvious due to other considerations. On the contrary, in view of the fact that strobilurins show the same mode of action with regard to ethylene production, one would expect it to be more likely to achieve an additive or even synergistic effect by combining with an ethylene modulator with a different mode of action.

The skilled person could also not expect that such compositions would be particularly suitable in processes for controlling rust in legumes, or for increasing yield and quality of legumes (also in the absence of pathogens), or for reducing ethylene evolution of plants, or for reducing undesired defoliation of crop plants, in that they show a stronger effect than the component alone, that one would have expected to have the particular effect, i.e. the strobilurin in case of controlling rust on legumes, the ethylene modulator in case of the other effects.

# Industrial Applicability

The subject-matter of claims 1-4, and 9-17 is considered to be industrially applicable (Article 33(1) and (4) PCT)

#### **Amended Claims:**

- 1. A mixture, comprising
- a) a compound of the formula I

in which

- x is halogen, C<sub>1</sub>-C<sub>4</sub>-alkyl or trifluoromethyl;
- 10 m is 0 or 1;
  - Q is C(=CH-CH<sub>3</sub>)-COOCH<sub>3</sub>, C(=CH-OCH<sub>3</sub>)-COOCH<sub>3</sub>, C(=N-OCH<sub>3</sub>)-COOCH<sub>3</sub> or N(-OCH<sub>3</sub>)-COOCH<sub>3</sub>;
    - A is -O-B, -CH<sub>2</sub>O-B, -OCH<sub>2</sub>-B, -CH=CH-B, -C=C-B, -CH<sub>2</sub>O-N=C(R<sup>1</sup>)-B or -CH<sub>2</sub>O-N=C(R<sup>1</sup>)-C(R<sup>2</sup>)=N-OR<sup>3</sup>, where
      - B is phenyl, naphthyl, 5-membered or 6-membered hetaryl or 5membered or 6-membered heterocyclyl which contains one to three nitrogen atoms and/or one oxygen or sulfur atom or one or two oxygen and/or sulfur atoms, where the ring systems are unsubstituted or substituted by one to three radicals R<sup>a</sup>:
        - is cyano, nitro, amino, aminocarbonyl, aminothiocarbonyl, halogen, C<sub>1</sub>-C<sub>6</sub>-alkyl, C<sub>1</sub>-C<sub>6</sub>-haloalkyl, C<sub>1</sub>-C<sub>6</sub>-alkylcarbonyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfonyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfoxyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-alkylsulfoxyl, C<sub>1</sub>-C<sub>6</sub>-alkylthio, C<sub>1</sub>-C<sub>6</sub>-alkylamino, di-C<sub>1</sub>-C<sub>6</sub>-alkylamino, C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, di-C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, C<sub>1</sub>-C<sub>6</sub>-alkylaminothiocarbonyl, di-C<sub>1</sub>-C<sub>6</sub>-alkylaminothiocarbonyl, C<sub>2</sub>-C<sub>6</sub>-alkenyl, C<sub>2</sub>-C<sub>6</sub>-alkenyloxy, phenyl, phenoxy, benzyl, benzyloxy, 5- or 6-membered heterocyclyl, 5- or 6-membered hetaryl, 5- or 6-membered hetaryloxy, C(=NOR')-OR" or OC(R')<sub>2</sub>-C(R")=NOR",

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where the cyclic radicals for their part are unsubstituted or substituted by one to three radicals R<sup>b</sup>:

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is cyano, nitro, halogen, amino, aminocarbonyl, aminothiocarbonyl, C<sub>1</sub>-C<sub>6</sub>-alkyl, C<sub>1</sub>-C<sub>6</sub>-haloalkyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfonyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfoxyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-alkoxycarbonyl, C<sub>1</sub>-C<sub>6</sub>-alkylthio, C<sub>1</sub>-C<sub>6</sub>-alkylamino, di-C<sub>1</sub>-C<sub>6</sub>-alkylamino, C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, di-C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, C<sub>1</sub>-C<sub>6</sub>-alkylaminothiocarbonyl, C<sub>2</sub>-C<sub>6</sub>-alkenyl, C<sub>2</sub>-C<sub>6</sub>-alkenyloxy, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, phenyl, phenoxy, phenylthio, benzyl, benzyloxy, 5- or 6-membered heterocyclyl, 5- or 6-membered hetaryloxy or C(=NOR')-OR";

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R' is hydrogen, cyano,  $C_1$ - $C_6$ -alkyl,  $C_3$ - $C_6$ -cycloalkyl or  $C_1$ - $C_4$ -haloalkyl;

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R" is hydrogen, C<sub>1</sub>-C<sub>6</sub>-alkyl, C<sub>3</sub>-C<sub>6</sub>-alkenyl, C<sub>3</sub>-C<sub>6</sub>-alkinyl, C<sub>1</sub>-C<sub>4</sub>-haloalkyl, C<sub>3</sub>-C<sub>6</sub>-haloalkenyl or C<sub>3</sub>-C<sub>6</sub>-haloalkinyl;

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R<sup>1</sup> is hydrogen, cyano, C<sub>1</sub>-C<sub>4</sub>-alkyl, C<sub>1</sub>-C<sub>4</sub>-haloalkyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, C<sub>1</sub>-C<sub>4</sub>-alkoxy;

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is phenyl, phenylcarbonyl, phenylsulfonyl, 5- or 6-membered hetaryl, 5- or 6-membered hetarylcarbonyl or 5- or 6-membered hetarylsulfonyl, where the ring systems are unsubstituted or substituted by one to three radicals R<sup>a</sup>,

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is  $C_1$ - $C_{10}$ -alkyl,  $C_3$ - $C_6$ -cycloalkyl,  $C_2$ - $C_{10}$ -alkenyl,  $C_2$ - $C_{10}$ -alkinyl,  $C_1$ - $C_{10}$ -alkylcarbonyl,  $C_2$ - $C_{10}$ -alkenylcarbonyl,  $C_3$ - $C_{10}$ -alkinylcarbonyl,  $C_1$ - $C_{10}$ -alkylsulfonyl or C(R')=NOR", where the hydrocarbon radicals of these groups are unsubstituted or substituted by one to three radicals  $R^c$ :

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is cyano, nitro, amino, aminocarbonyl, aminothiocarbonyl, halogen, C<sub>1</sub>-C<sub>6</sub>-alkyl, C<sub>1</sub>-C<sub>6</sub>-haloalkyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfonyl, C<sub>1</sub>-C<sub>6</sub>-alkylsulfoxyl, C<sub>1</sub>-C<sub>6</sub>-alkoxy, C<sub>1</sub>-C<sub>6</sub>-haloalkoxy, C<sub>1</sub>-C<sub>6</sub>-alkylsulfoxyl, C<sub>1</sub>-C<sub>6</sub>-alkylthio, C<sub>1</sub>-C<sub>6</sub>-alkylamino, di-C<sub>1</sub>-C<sub>6</sub>-alkylamino, C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, di-C<sub>1</sub>-C<sub>6</sub>-alkylaminocarbonyl, di-C<sub>1</sub>-C<sub>6</sub>

alkylaminothiocarbonyl, C<sub>2</sub>-C<sub>6</sub>-alkenyl, C<sub>2</sub>-C<sub>6</sub>-alkenyloxy, C<sub>3</sub>-C<sub>6</sub>-cycloalkyl, C<sub>3</sub>-C<sub>6</sub>-cycloalkyloxy, 5- or 6-membered heterocyclyl, 5- or 6-membered heterocyclyloxy, benzyl, benzyloxy, phenyl, phenoxy, phenylthio, 5- or 6-membered hetaryl, 5- or 6-membered hetaryloxy or hetarylthio, where the cyclic groups for their part may be partially of fully halogenated or may carry one to three radicals R<sup>a</sup>; and

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 $R^3$  is hydrogen,  $C_1$ - $C_6$ -alkyl,  $C_2$ - $C_6$ -alkenyl or  $C_2$ - $C_6$ -alkinyl, where the hydrocarbon radicals of these groups may be unsubstituted or substituted by one to three radicals  $R^c$ ;

and

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- b) one or more ethylene modulators (II) selected from the group consisting of:
  - ethylene biosynthesis inhibitors which inhibit the conversion of Sadenosyl-L-methionine into 1-aminocyclopropane-1-carboxylic acid (ACC), selected from derivatives of vinylglycine and hydroxylamines;

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o ethylene biosynthesis inhibitors which block the conversion of ACC into ethylene, selected from the group consisting of: Co<sup>++</sup> or Ni<sup>++</sup> ions in plant-available forms; phenolic radical scavengers such as *n*-propyl gallate; polyamines, such as putrescine, spermine or spermidine; structural analogs of ACC, such as α-aminoisobutyric acid or L-aminocyclopropene-1-carboxylic acid; salicylic acid or acibenzolar-S-methyl; structural analogs of ascorbic acid which act as inhibitors of ACC oxidase, such as prohexadione-Ca or trinexapac-ethyl; and triazolyl compounds such as paclobutrazol or uniconazole as inhibitors of

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o inhibitors of the action of ethylene selected from the group consisting of: structural analogs of ethylene such as 1-methylcyclopropene or 2,5-norbornadiene and 3-amino-1,2,4-triazole or Ag<sup>++</sup> ions

cytochrome P-450-dependent monooxygenases whose main action is

in a weight ratio of I to II of from 20:1 to 0.05:1.

to block the biosynthesis of gibberellins;

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2. A mixture as claimed in claim 1 where the compound of the formula I is a stro-bilurin derivative selected from the group consisting of azoxystrobin, dimoxy-strobin, fluoxastrobin, kresoxim-methyl, metominostrobin, orysastrobin, trifloxy-strobin, picoxystrobin or pyraclostrobin.

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- 3. A mixture as claimed in claim 1 where the compound of the formula I is pyraclostrobin.
- 4. A mixture as claimed in claim 1 where the ethylene modulators are Co<sup>++</sup> ions, aminoethoxyvinylglycine, aminooxyacetic acid, prohexadione-Ca, trinexapacethyl, α-aminoisobutyric acid, salicylic acid or 3-amino-1,2,4-triazole.
  - 5. A mixture as claimed in claim 1 where the ethylene modulators are Co<sup>++</sup> ions.
- 10 6. A mixture as claimed in claim 1 where the ethylene modulators is prohexadione-
  - 7. A mixture as claimed in claim 1 where the ethylene modulator is salicylic acid.
- 15 8. A mixture as claimed in claim 1 where the ethylene modulators are prohexadione-Ca together with Co<sup>++</sup> ions.
- 9. A mixture as claimed in any of claims 1 to 8 which additionally comprises an azole III selected from the group consisting of bromoconazole, cyproconazole, epoxiconazole, fenbuconazole, fluquiconazole, flusilazole, metconazole, myclobutanil, propiconazole, prochloraz, prothioconazole, tebuconazole or triticonazole.
- 10. A mixture as claimed in any of claims 1 to 9 which additionally comprises a surfactant selected from the group consisting of: polyoxyethylene sorbitan monolaurate,
   25 alkylphenoxy polyethoxy ethanol, fatty alcohol, fatty alcohol alkoxylate and sodium dodecylsulfate.
- 11. A method for controlling rust infections in legumes, which comprises treating the above-ground plant parts of the legumes with an aqueous preparation of a mixture as claimed in any of claims 1 to 10.
  - 12. A process as claimed in claim 11, wherein rust infection on leaves and fruits of soya plants is controlled.
- 13. A process as claimed in claim 11, wherein the rust infection is caused by *Phakop-sora* pachyrhizi and/or *Phakopsora meibomiae*.
  - 14. A process for increasing the yield and quality of legumes by using mixtures as claimed in any of claims 1 to 10.

15. A method for increasing the yield and quality of legumes applying an effective amount of a mixture as claimed in any of claims 1 to 10.

- 16. A method for reducing the ethylene evolution of plants by applying an effective amount of a mixture as claimed in claims 1 to 10.
  - 17. A method for reducing undesired defoliation of crop plants by applying an effective amount of a mixture as claimed in claims 1 to 10.
- 10 18. A method for controlling harmful plant pathogens by applying an effective amount of Co<sup>++</sup> ions in plant-available form.

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